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CONTROL AND SYNCHRONIZATION OF DYNAMICAL SYSTEMS:
STUDIES OF CHEMICAL MODEL SYSTEMS

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Studies of Chemical Model Systems
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FINAL Report - Part I

- a. Submitted Papers to Refereed Journals (not yet published): 1
- b. Published Papers in Refereed Journals: 8
1. E. Mihaliuk, R. Wackerbauer, and K. Showalter, "Topographic Organization of Hebbian Neural Connections by Synchronous Wave Activity," *Chaos*, **11**, 287-292 (2001).
 2. M. Hildebrand, H. Skødt, and K. Showalter, "Spatial Symmetry Breaking in the Belousov-Zhabotinsky Reaction with Light-Induced Communication," *Phys. Rev. Lett.* **87**, 883031-4 (2001).
 3. E. Mihaliuk, T. Sakurai, F. Chirila, and K. Showalter, "Experimental and Theoretical Studies of Feedback Stabilization of Propagating Wave Segments, *Faraday Discussions*, *Roy. Chem. Soc.* **120**, 383-394 (2002).
 4. E. Mihaliuk, T. Sakurai, F. Chirila, and K. Showalter, "Feedback Stabilization of Unstable Waves," *Phys. Rev. E* **65**, 656021-4 (2002).
 5. T. Sakurai, E. Mihaliuk, F. Chirila, and K. Showalter, "Design and Control Patterns of Wave Propagation Patterns in Excitable Media," *Science* **296**, 2009-2012 (2002).

6. M. Hildebrand, J. Cui, E. Mihaliuk, J. Wang, and K. Showalter, "Synchronization of Spatiotemporal Patterns in Locally Coupled Excitable Media," *Phys. Rev. E* **68**, 0262051-4 (2003).
 7. R. Wackerbauer and K. Showalter, "Collapse of Spatiotemporal Chaos," *Phys. Rev. Lett.* **91**, 1741031-4 (2003).
 8. J. Wang, H. Sun, S. K. Scott, and K. Showalter, "Uncertain Dynamics in Nonlinear Chemical Reactions," *Phys. Chem. Chem. Phys.* **5**, 5444-5447 (2003).
- c. Submitted Book Chapters (not yet published): 0
 - d. Published Book Chapters: 0
 - e. Printed Non-refereed Technical Reports: 0
 - f. Patents Filed: 0
 - g. Patents Granted: 0
 - h. Invited Presentations: 30
 1. "Periodic Driving and Feedback Stabilization of Wave Segments." Sixth SIAM Conference on Applications of Dynamical Systems; Snowbird, Utah; May 19-25, 2001.
 2. "Periodic Driving and Feedback Stabilization of Chemical Waves." Experimental Chaos Conference; Potsdam, Germany; July 22-26, 2001.
 3. "Feedback Stabilization of Propagating Wave Segments." Patterns and Waves - Mathematics and Nonlinear Chemistry; Lorentz Center, Leiden University; Leiden, The Netherlands; September 1-7, 2001.
 4. "Feedback Stabilization of Propagating Wave Segments." Faraday Discussions 120; Manchester, England; September 10-12, 2001.
 5. "Perturbed Excitable Media." Federation of Analytical Chemistry and Spectroscopy Societies, Symposium on Electrochemical Oscillations and Nonlinear Systems; Detroit, Michigan; October 9, 2001.

6. "Chemical Waves and Patterns." Departmental Seminar, Facultad de Ciencias, Universidad Autónoma del Estado de Morelos, Cuernavaca, Morelos, México; May 13, 2002.
7. "Feedback Stabilization of Propagating Waves." Conference on Engineering of Chemical Complexity, Fritz-Haber-Institut, Berlin, Germany; June 5-7, 2002.
8. "Chemical Waves and Patterns." Departmental Seminar, Institute for Normal and Abnormal Physiology, University of Marburg, Marburg, Germany; June 20, 2002.
9. "Feedback Stabilization of Propagating Waves." School on Fundamentals and Perspectives of Nonlinear Dynamics, International Centre of Condensed Matter Physics, University of Brasilia, Brasilia, Brazil; July 1-5, 2002.
10. "Spatiotemporal Chaos in a Chemical Reaction-Diffusion System: Competitive Chaotic Populations and Transient Dynamics." School on Fundamentals and Perspectives of Nonlinear Dynamics, International Centre of Condensed Matter Physics, University of Brasilia, Brasilia, Brazil; July 1-5, 2002.
11. "Spatiotemporal Chaos in a Chemically Realistic Model: Dynamics, Competitive Chaotic Populations and Transient Behavior." School and Workshop on Spatiotemporal Chaos, International Centre for Theoretical Physics, Trieste, Italy; July 14-21, 2002.
12. "Detailed Characterization of the Excitability Limit for Spiral Waves," Gordon Research Conference on Oscillations and Dynamic Instabilities in Chemical Systems, Queens College, Oxford University, Oxford, U.K.; July 23-August 2, 2002.
13. "Chemical Waves and Patterns," Department of Biochemistry and Molecular Pharmacology Colloquium, West Virginia University, Morgantown, WV; October 7, 2002.
14. "Chemical Waves and Patterns," Department of Physics Colloquium, West Virginia University, Morgantown, WV; October 10, 2002.
15. "Chemical Waves and Patterns." Department of Physics Colloquium, Indiana University of Pennsylvania; April 17, 2003.
16. "Chemical Waves and Patterns." Department of Chemistry and Biochemistry Colloquium, University of Windsor, Windsor, Ontario; April 25, 2003.

17. "Feedback Stabilization and Control of Unstable Propagating Waves." 2003 SIAM Conference on Applications of Dynamical Systems, Snowbird, Utah; May 26-31, 2003.
18. "Spatial Symmetry Breaking with Light-Induced Remote Communication." 2003 SIAM Conference on Applications of Dynamical Systems, Snowbird, Utah; May 26-31, 2003.
19. "Chemical Waves and Patterns." Departments of Applied Mathematics and Chemical Engineering Joint Colloquium, Northwestern University, Evanston, Illinois; January 16, 2003.
20. "Stabilization and Control of Unstable Propagating Waves." Symposium on Pattern Formation and Stochastic Processes in Biological Systems, Institute of Physics, Humboldt University, Berlin, Germany; June 30, 2003.
21. "Stabilizing and Controlling Unstable Propagating Waves". Max-Planck-Institute Sonderforschungsbereich Symposium, Berlin, Germany; July 11, 2003.
22. "Feedback Stabilization and Control of Unstable Propagating Waves." Second International Symposium on Molecular Synchronization for Design of New Materials, Tokyo Institute of Technology, Yokohama, Japan; July 18-19, 2003.
23. "Stabilization and Control of Unstable Propagating Waves." Gordon Research Conference on Nonlinear Science, Tilton, New Hampshire; August 2-8, 2003.
24. "Stabilization and Control of Unstable Propagating Waves." Conference on Trends in Pattern Formation: From Amplitude Equations to Applications, Max-Planck-Institut Physik für Komplex Systems, Dresden, Germany; September 15-19, 2003.
25. "Controlling Spatiotemporal Dynamics in Chemical Systems." Department of Chemical Engineering Colloquium, University of Virginia, Charlottesville, Virginia; October 30, 2003.
26. "Spatiotemporal Chaos in a Chemical Model: Dynamics, Competitive Chaotic Populations and Transient Behavior." Research Institute for Electronic Science Colloquium, Hokkaido University, Sapporo, Japan; November 28, 2003
27. "Feedback Stabilization and Control of Unstable Propagating Waves." Fifth RIES-Hokudai International Symposium, Hokkaido University, Sapporo, Japan; December 1-2, 2003.

28. "Addressable Excitable for Modeling Population Dynamics." Centro de Ciencias Fisicas, Universidad Nacional Autónoma de México (UNAM), Cuernavaca, Mexico; February 25, 2004.
 29. "Modeling Disease Spreading and Collective Behavior Using Addressable Excitable Media." Vice President's Distinguished Lecture, Departments of Physics, University of Missouri at Saint Louis and University of Missouri at Rolla; March 5, 2004.
 30. "Stabilization and Control of Unstable Propagating Waves." Pattern Formation and Collective Phenomena Focus Session, March American Physical Society Meeting, Montreal, Canada; March 23, 2004.
- i. Submitted Presentations: 0
- j. Honors/Awards/Prizes:
1. Alexander von Humboldt Senior Scientist Award, 1999-2004.
 2. Co-Chair, Gordon Research Conference on Nonlinear Science, 2001.
- k. Full-time equivalent Graduate Students and Post-Doctoral Associates: 6
- Graduate Students: 4
- Post-Doctoral Associates: 2
- Female Graduate Students: 2
- Female Post-Doctoral Associates: 0
- Minority* Graduate Students: 1
- Minority* Post-Doctoral Associates: 0
- Asian Graduate Students: 2
- Asian Post-Doctoral Associates: 1

1. Other Funding

1. National Science Foundation, "Spatiotemporal Dynamics in Chemical Systems," current year funding: \$0, total funding: \$341,589, grant period: April 1999 - June 2002. This project involved studies of inhomogeneous excitable media, pattern formation, and applications of control theory to excitable media.
2. National Science Foundation, "Spatiotemporal Dynamics in Chemical Systems," current year funding: \$0, total funding: \$259,000, grant period: July 2002 - June 2004. This grant was a National Science Foundation "Two-Year Extension for Special Creativity" of my previous NSF grant that expired June 2002. The project involved studies of inhomogeneous excitable media, pattern formation, and applications of control theory to chemical systems.

FINAL Report - Part II

- a. Principal Investigator: Kenneth Showalter
- b. Phone Number: (304) 293-3435 x6428
- c. Cognizant ONR Program Officer: Michael Shlesinger
- d. Program objective: The development and application of control methods for spatiotemporal systems.
- e. Significant results:

In Ref. 1 (see Part I, b.), "Topographic Organization of Hebbian Neural Connections by Synchronous Wave Activity," we study the evolution of initially random unidirectional connections between two excitable layers of FitzHugh-Nagumo neurons with simulated spontaneous activity in the input layer. We find in this simple model system a rapid topographic reorganization arising from synchronous wave activity due to lateral diffusive coupling within the layers and from Hebbian learning in the coupling between the layers. Topographically organized patterns of neural connections in which neighboring neurons project to neighboring sites in the target occur throughout the nervous system. In the vertebrate visual system, the retina sends images to be further processed in the lateral geniculate nucleus (LGN). The required topographically precise connection network develops by processes that are not fully understood but are believed to involve electrical activity in the retina before the onset of vision.

In Ref. 2, "Spatial Symmetry Breaking in the Belousov-Zhabotinsky Reaction with Light-Induced Communication," we report on novel spatiotemporal patterns in the photosensitive BZ reaction arising from a nonlocal feedback that imposes short-range activation and long-range inhibition. Domains containing spiral waves form on a stationary background with light-induced feedback that alternates from positive (activatory) for short distances to negative (inhibitory) for larger distances. Complex behavior of colliding and splitting wave fragments is found with feedback radii comparable to the spiral wave length. We show that such a feedback gives rise to a "Turing-like" instability, and, as a result, patterns with more than one characteristic length scale are formed.

We present a detailed characterization of the excitability limit for spiral waves in Ref. 3,

"Experimental and Theoretical Studies of Feedback Stabilization of Propagating Wave Segments."

The evolution of waves with free ends is influenced not only by the excitability of the medium but also by the wave size, such that smaller wave segments contract laterally until they disappear while larger waves expand to form spirals. There is a critical wave size separating these two outcomes that is an unstable waveform inherent to the medium, which will either grow or decay when perturbed. Different critical wave sizes exist for different excitabilities in the boundary regime, and the size of these waves increases with decreasing excitability. We study the locus of critical wave sizes by stabilizing the unstable waves with a feedback control algorithm. The medium excitability is continually adjusted according to the wave area such that it is increased when the wave area decreases and vice versa, thereby stabilizing the wave segment at its critical size. We describe experimental and numerical studies implementing this control method in the Belousov-Zhabotinsky reaction and present a theoretical study based on a kinematic analysis.

In Ref. 4, "Feedback Stabilization of Unstable Waves," we refine our theoretical description of stabilized unstable waves in weakly excitable media. The locus of steady-state wave size as a function of excitability defines the perturbation threshold for the initiation of spiral waves. This locus also defines the excitability boundary for spiral wave behavior in active media. Our kinematic analysis, based on stationarity of the stabilized unstable waves and velocity dependence on curvature, yields the wave shape and size as a function of excitability. The analysis is generally applicable to all excitable media.

Intricate patterns of wave propagation are exhibited in a chemical reaction-diffusion system with spatiotemporal feedback. In Ref. 5, we describe particle-like waves that propagate in effectively user defined patterns. Normally unstable, the waves are first stabilized by global feedback to the overall excitability of the medium. The wave motion is controlled by a secondary feedback to yield a desired trajectory, such as hypotrochoid target trajectories. Wave propagation akin to Brownian motion is exhibited when random variations in the excitability gradient are imposed. The variations lead to completely different explorations of the medium by subsequent waves. When a boundary rule is incorporated into the control algorithm, waves are confined to a designated region of the medium. A wave can be confined to the vicinity of a rectangular box by imposing a constant gradient outside the box, maintained perpendicular to the direction of wave propagation, which causes it to reenter

the box. Two waves "interacting" with each other are observed when attraction and repulsion terms are incorporated into the control algorithm. When the interaction strength is the same for both waves, they settle into a stable orbit with their separation maximized. Our experiments and simulations demonstrate how different modes of feedback can be combined to give patterns of spatiotemporal behavior that would otherwise not be possible. Spatiotemporal behavior is pervasive in living systems, and these control methods offer a promising direction for probing underlying mechanisms.

The synchronization of coupled chaotic oscillators has attracted much attention in recent years, and complete, phase, lag, and generalized synchronization have been distinguished in such systems. Recently, synchronization phenomena in spatially extended systems have attracted increasing attention, and identical synchronization and phase synchronization have been observed in systems exhibiting spatiotemporal chaos. The synchronization of two distributed Belousov-Zhabotinsky systems is experimentally and theoretically investigated in Ref. 6, where symmetric local coupling is carried out using a video camera-projector setup. Spatial disorder in the coupled systems, introduced as random initial configurations of spirals, gradually decreases until a final state is attained that corresponds to a synchronized state with a single spiral in each system. The experimental observations are confirmed with numerical simulations of two identical Oregonator models with symmetric local coupling, and a systematic study revealed generalized synchronization of spiral waves.

In Ref. 7, the transient nature of spatiotemporal chaos is examined in reaction-diffusion systems with coexisting stable states. Transient spatiotemporal chaos has been suggested as an explanation for species extinction in ecological systems, and it has been studied in models for semi-conductor charge transport and the CO oxidation on single-crystal Pt surfaces. Chaos-like spatiotemporal dynamics has also been found to be transient in a system of coupled one-dimensional maps. We find the apparent asymptotic spatiotemporal chaos of the Gray-Scott system to be transient, with the average transient lifetime increasing exponentially with medium size. The collapse of spatiotemporal chaos arises when statistical spatial correlations produce a quasi-homogeneous medium, and the system obeys its zero-dimensional dynamics to relax to its only stable asymptotic state.

Complex chemical behavior, such as periodic, quasiperiodic, and chaotic oscillations, has

been widely studied, e.g., in continuous-flow stirred tank reactors and as transients in batch reactors. Such dynamical behavior has challenged long-held notions concerning reproducibility in chemical systems that have the same experimental conditions. In Ref. 8, we study how different initial conditions can give rise to completely different dynamics in a chemical model system, from steady state to periodic to chaotic behavior. We describe how deterministic chemical kinetics can exhibit an extreme sensitivity to fluctuations, such that qualitatively different dynamical behaviors would be explored in an experimental chemical system where such fluctuations are unavoidable.

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14. ABSTRACT <p>Studies of spatiotemporal dynamical behavior in chemical systems were carried out. We have built on our expertise in addressable excitable media to develop and characterize particle-like waves that autonomously navigate and traverse obstacle-filled landscapes. Experimental investigations have been combined with theoretical and computational analyses in each project. The results of our studies are generally applicable, and our advances in designing and controlling spatiotemporal dynamical systems will be beneficial to the Department of Defense.</p>					
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